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High-Triplet-Energy Bipolar Host Materials Based on Phosphine Oxide Derivatives for Efficient Sky-Blue Thermally Activated Delayed Fluorescence Organic Light-Emitting Diodes with Reduced Roll-off

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We designed and synthesized two new ambipolar host materials, namely **CzPO** and **Cz3PO**, which contain electron-donating carbazole and electron-accepting triphenylphosphine oxide moieties. Thermally activated delayed fluorescence (TADF)-based OLEDs employing **CzPO** and **Cz3PO** as host materials and the 9-(4-(4,6-diphenyl-1,3,5-triazin-2-yl)phenyl)-1,3,6,8-tetramethyl-9H-carbazole (**CzTRZ2**) as the emitter resulted in improved maximum external quantum efficiencies, EQE_{max}, of 13.1% and 13.2%, respectively, together with small efficiency roll-offs, while the device based on bis[2-(diphenylphosphino)phenyl]ether oxide (**DPEPO**) showed a much more pronounced efficiency roll-off. The reduced efficiency roll-off in the devices based on **CzPO** and **Cz3PO** can be ascribed to their improved ambipolar charge transport capacity compared to that of **DPEPO**, which results in a broader recombination zone in the emitting layer using these ambipolar hosts.

Keywords: Thermally activated delayed fluorescence (TADF) | Organic light-emitting diode (OLED) | Ambipolar host

After the invention of fluorescence-based organic light-emitting diodes (OLEDs) in 1987[1], the flat panel display market based on OLEDs is emerging to become the dominant technology. Both organic-based fluorescence and organometallic-based phosphorescence emitters are currently used in OLEDs[2,3]. In 2012, a series of highly efficient OLEDs based on thermally activated delayed fluorescence (TADF) were demonstrated, in which an efficient process of reverse intersystem crossing (RISC) from a triplet excited-state to a singlet excited-state was realized in purely organic molecules[4]. OLEDs based on TADF emitters achieved comparable an internal quantum efficiency (IQE) of 100%. Since then, a large number of TADF emitters have been reported[5]. In particular, TADF emitters have shown tremendous potential to replace the state-of-the-art fluorophores in high performance deep-blue OLEDs. During the survey of blue-emitting devices, significant efficiency roll-off at high current density was identified in these devices, and the triplet energies (E_T) of the host materials must be sufficiently high compared to the emitters in order to confine the excitons on the emitters and thereby realize efficient blue OLEDs[5c]. Further, the balance of charge transport carriers must also be considered to mitigate the efficiency roll-off.

For blue TADF-based OLEDs, there are some general requirements for the host material: (1) the E_T of host materials must be higher than that of the emitter to prevent back energy transfer[6]; (2) to facilitate effective charge injection, both the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) energy levels of the host materials should be well matched with both those of the emitter and the other layers of the device[7]; (3) a good host material should possess ambipolar character to best manage charge transport through the emissive layer[8,9]. In general, the recombination region tends to occur closer to the emissive layer/electron-transporting layer (EML/ETL) interface when using hole-transporting (HT) hosts. Likewise, the recombination region is likely to be located in the EML/HTL interface when using an electron-transporting (ET) host such as the frequently used DPEPO. It has been found that poor and unbalanced carrier mobility within the EML is responsible for inferior efficiencies, low stability and efficiency roll-off[10]. OLEDs with narrow charge recombination zones lead to devices presenting severe efficiency roll-off due to the local accumulation of high-density triplet excitons, especially at high current densities[11-15]. As a result, an ambipolar host is more suitable for improving the device performance in blue OLEDs. The simple incorporation of both HT and ET moieties within host materials should give rise to ambipolar host materials[16-19]. **DPEPO** is the most popular host material for blue-emitting OLEDs[20]. However, the use of such ET-type host materials is usually coupled with reduced device stability and efficiency roll-off[21-24]. Thus, in this study, we designed two host materials, **CzPO** and

Cz3PO, by linking carbazole donor units with an electron-accepting triphenylphosphine oxide group (Fig. 1). Both hosts show high E_Ts and ambipolar charge transport properties. Sky-blue OLEDs based on these two bipolar host materials not only showed smaller efficiency roll-off characteristics but also operated under relatively low driving voltages.

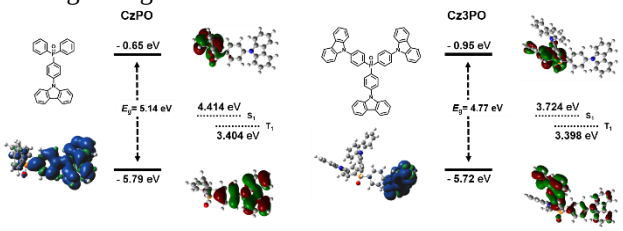

Figure 1. Molecular structures, HOMO, LUMO, S_1 and T_1 energy levels of **CzPO** and **Cz3PO** characterized by DFT calculations at the PBE0/6-31G(d) level of theory, and the electron density distributions of the frontier molecular orbitals and spin density distributions of T_1 energy levels.

The energy levels of **CzPO** and **Cz3PO** were first investigated computationally using the Gaussian 16 program package[25]. Ground-state geometries in the gas phase were optimized at the PBE0/6-31G(d) level, and the lowest singlet and triplet excited states were calculated using time-dependent density functional theory (TD-DFT) based on the optimized ground-state geometries. The electron density distribution of the HOMOs and LUMOs of **CzPO** and **Cz3PO** are depicted in Fig. 1 along with their energies and the energies of the singlet and triplet excited states and the spin density distribution of the T_1 states. The HOMOs of both host materials are delocalized over the electron-donating carbazole units and bridging phenylene while the LUMOs are mainly localized over the electron-accepting phosphine oxide moiety.

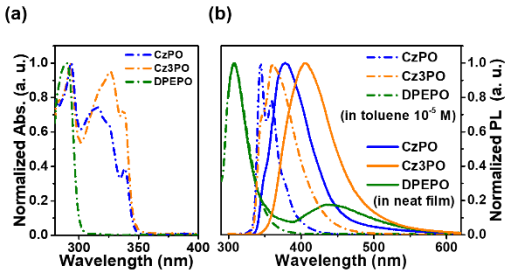

Figure 2. UV-Vis absorption spectra of **CzPO**, **Cz3PO** and **DPEPO** in toluene (10^{-5} M) and (b) PL emission spectra of **CzPO**, **Cz3PO** and **DPEPO** in toluene (10^{-5} M) and neat films.

The UV–Vis absorption and photoluminescence (PL) spectra of **CzPO**, **Cz3PO** and **DPEPO** are shown in Fig. 2 respectively. The photophysical data are summarized in Table S1. In toluene, all host materials show strong absorption bands below 300 nm, which are assigned as π–π* transitions of the phosphine oxide moiety. The absorption band beyond 300 nm is absent in **DPEPO**, and the lowest-energy absorption bands ranging from 310 to 350 nm are attributed to the π–π* transition of the 9-phenylcarbazole unit in **CzPO** and **Cz3PO**. In toluene, **CzPO** shows emission maxima (λ_{PL}) at 344 and 359 nm, while **Cz3PO** shows λ_{PL} at 361 nm. DPEPO shows the most hypsochromically shifted emission at 290 nm. The bathochromically shifted emission in **Cz3PO** is due to the presence of three donor groups within the molecule. In neat films, only **CzPO** and **Cz3PO** display red-shifted emission at 377 and 406 nm, respectively, whereas **DPEPO** show an additional emission band at 437 nm that may be ascribed to excimer emission.[28] In the present study, **CzTRZ2** was used as the TADF emitter in the emitting layer as the absorption spectrum of **CzTRZ2** overlaps strongly with the neat film emission spectra of **CzPO** and **Cz3PO**, thereby assuring efficient Förster resonance energy transfer (FRET) from the excited hosts to **CzTRZ2**.

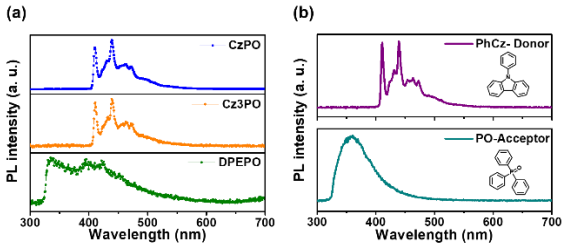

Figure 3. (a) Phosphorescence spectra of **CzPO**, **Cz3PO** and **DPEPO** in toluene matrices at 77 K. (b) Phosphorescence spectra of donor and acceptor moieties in toluene matrices at 77 K.

The phosphorescence spectra of **CzPO**, **Cz3PO** and **DPEPO** are depicted in Fig. 3a and these are cross-compared with the phosphorescence spectra of *N*-phenylcarbazole and triphenylphosphine oxide (Fig. 3b), which act as reference electron-donor and electron-acceptor moieties. From the phosphorescence spectra, the ET values were determined to be 3.06 and 3.05 eV for **CzPO** and **Cz3PO**, respectively. The E_T of **CzPO** and **Cz3PO** are higher than that of **CzTRZ2** (2.85 eV)[5d], ensuring a suppression of back energy transfer. Both **CzPO** and **Cz3PO** show similar vibronic-structured emission profiles and E_T to that of *N*-phenylcarbazole. Therefore, the lowest T_1 excited state of each of **CzPO** and **Cz3PO** is localized on the donor moieties, consistent with the computed spin density simulation.

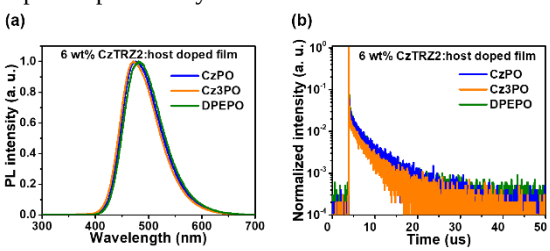

Figure 4. (a) Steady-state PL spectra and (b) time-resolved PL decay profiles of 6 wt.%-CzTRZ2:host doped thin films (host = **CzPO**, **Cz3PO**, and **DPEPO**) measured at 300 K under N_2 . (excitation wavelength: 280 nm).

The PL spectra of 6 wt %-**CzTRZ2** doped films in **CzPO**, **Cz3PO** and **DPEPO** are nearly identical, indicating efficient FRET (Fig. 4a). The **CzPO**-, **Cz3PO**- and **DPEPO**-doped films of **CzTRZ2** show broad and structureless sky-blue emission with high Φ_{PL} of 81%, 75%, and 92%, respectively. The transient decay profiles of the doped films clearly show biexponential decay respectively, at 300 K (Figs. S1 and 4b). Such behavior is characteristic of the TADF emission previously reported for this compound.[5d] Furthermore, both hosts materials possess a high decomposition temperature, T_d, of 365 and 454 °C, for **CzPO** and **Cz3PO**, respectively, which is indicative of high thermal and morphological stabilities (Fig. S2). The T_d values are substantially higher than that of **DPEPO** (T_d = 322 °C)[26].

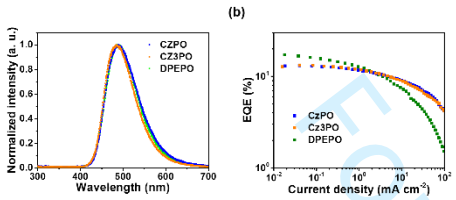

Figure 5. (a) The EL spectra measured at 8 V, and (b) external quantum efficiency (η_{ext})–current density curves of the TADF-based OLEDs.

We next evaluated the EL performance in OLEDs using **CzTRZ2** as the TADF emitter and **CzPO**- and **Cz3PO** and **DPEPO** as host matrices in the EML. The devices were fabricated with the following architecture : ITO (100 nm)/HATCN (10 nm)/TAPC (40 nm)/mCP (10 nm)/6 wt.%-CzTRZ2:host (20 nm)/PPT (10 nm)/TPBi (50 nm)/LiF (0.8 nm)/Al (100 nm), in which 2,3,6,7,10,11-hexacyano-1,4,5,8,9,12-hexaazatriphenylene (HATCN) is the hole-injection layer, 1,1-bis(4-ditolylaminophenyl) cyclohexane (TAPC) and 1,3-bis(*N*-carbazolyl)benzene (mCP) are the hole-transporting layer and electron-blocking layer, respectively. The EML layer consists of 6 wt.% of **CzTRZ2** doped in **CzPO**, **Cz3PO** or **DPEPO**. 2,8-Bis(diphenylphosphoryl)dibenzothiophene (PPT) is the hole-blocking layer and 1,3,5-tris(*N*-phenylbenzimidazol-2-yl)benzene (TPBi), LiF and Al serve as the electron-transporting layer, the electron-injection layer and cathode, respectively. The ET of electron-blocking and hole-blocking layers, i. e., mCP (2.9 eV) and PPT (2.9 eV), respectively, are higher than that of **CzTRZ2**, which ensures confinement of the triplet excitons within the EML[27]. The EL spectra and EQE characteristics of the corresponding OLEDs are shown in Fig. 5. Devices based on **CzPO** and **Cz3PO** as host materials resulted in high EQE_{max} of 13.1 and 13.2% respectively. By contrast, the device based on DPEPO showed a higher EQE_{max} of 16.7%. Despite the higher EQE_{max}, the **DPEPO**-based devices showed a significantly serious efficiency roll-off with an EQE of only 1% at 100 mA cm^{-2}. Meanwhile, the **CzPO**- and **Cz3PO**-based devices exhibited reduced efficiency roll-off with EQE_{100} of 4.2% and 4.2%, respectively, at a display-relevant current density of 100 mA cm^{-2}. All the device data are shown in Table S2.

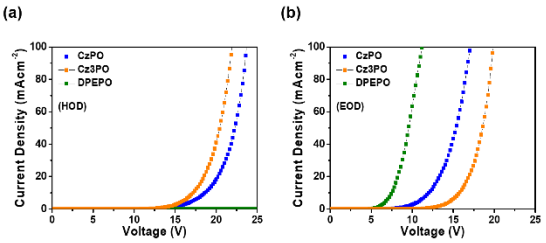

Figure 6. Current density–voltage (*J*–*V*) characteristics of (a) hole-only devices (HODs) and (b) electron-only devices (EODs) based on the **CzPO**, **Cz3PO** and **DPEPO** hosts.

To investigate the reason for the reduced roll-off characteristics, hole-only devices (HODs) and electron-only devices (EODs) with different host materials were fabricated to evaluate the carrier transporting properties of the corresponding OLEDs. These device configurations were: ITO/HAT-CN (10 nm)/TAPC (20 nm)/ host (60 nm)/TAPC (20 nm)/Al (100 nm) for the HODs and ITO/TPBi (20 nm)/host (60 nm)/TPBi (20 nm)/LiF (0.8 nm)/Al (100 nm) for the EODs. Figure 6 showed the *J*–*V* curves of the fabricated HODs and EODs. For the HODs (Fig. 6a), the **Cz3PO** and **Cz3PO**-based devices showed hole transporting/injection properties due to the presence of more electron-donating carbazole units, while no significant electron current was observed in **DPEPO**-based device. In contrast, for EODs, the order of the electron transporting/injection properties of devices was **DPEPO** > **CzPO** > **Cz3PO** (Fig. 6b). From both the HODs and EODs characteristics, **CzPO** and **Cz3PO** were confirmed to be ambipolar in nature, in which **CzPO**- and **Cz3PO**-based devices exhibited balanced hole and electron transporting/injection properties. Thus, the well-balanced charge flux and broad distribution of the charge-recombination zones within the EML resulted in low efficiency roll-off characteristics for **CzPO**- and **Cz3PO**-based OLEDs.

In summary, reduced efficiency roll-offs were demonstrated in sky blue TADF-based OLEDs using **CzPO** and **Cz3PO** as ambipolar hosts compared to the device with the reference **DPEPO** host. Sky blue TADF-OLEDs based employing **CzPO** and **Cz3PO** host materials showed maximum EQEs of 13.1 and 13.2%, respectively, and reduced roll-offs with EQEs of 4.2% and 4.2%, respectively, at current densities of 100 mA cm^{-2}.

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Graphical Abstract

Textual Information

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Graphical Information

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